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HIGHLIGHTS REPORT FOR FISSION PRODUCT RELEASE TEST VI-1

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1. INTRODUCTION

This report summarizes the preliminary findings of the first of a series of fission product release tests conducted in a rebuilt experimental facility. The work, sponsored by the U.S. Nuclear Regulatory Commission (NRC), is a continuation of fission product release studies which have been conducted at Oak Ridge National Laboratory (ORNL) for several years. This work has emphasized studies of commercial LWR fuel. All previous tests in this fission product release project were conducted in horizontally oriented furnaces.[1,2] This test, however, is the first in a series of tests in a vertical furnace and collection apparatus.

2. PURPOSE OF TEST

The purpose of test VI-1 was to heat a vertically mounted specimen of Oconee reactor fuel for periods of 20 min each at temperatures of 2000 and 2300 K in steam flowing at ~2 L/min. The first temperature level duplicated test HI-5, except for the higher steam flow rate in the later test, and also duplicated test HI-2, except that it used H. B. Robinson fuel.[3,4] The second phase provided release data at a higher temperature, thus permitting comparison of release at two different constant temperatures from the same fuel specimen. The higher steam flow rate was designed to effect complete oxidation of the Zircaloy cladding early in the test, so that no clad melting would occur at the higher temperature. Because reactor fuel rods are mounted vertically, the vertical orientation of the new furnace should provide a more realistic environment for tests with clad melting, but no significant differences, compared with the horizontal tests, were expected in this case. The use of three different fission product collection trains (A, B, and C) operated in series provided data for three different time periods during the test.

3. DESCRIPTION OF APPARATUS

The current test apparatus[5] is illustrated in Fig. 1, and details of furnace construction are shown in Fig. 2. In addition to the vertical furnace, the principal features of the new apparatus are (1) three collection trains rather than one; (2) temperature measurement via windows in the side of the furnace, with optical fibers or a mirror conducting the light to pyrometers mounted outside the hot cell; and (3) equipment mounted downstream from the cold charcoal traps to continuously measure the production of hydrogen during the test. The thermocouple mounted in

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*Guest scientist from the Japan Atomic Energy Research Institute.

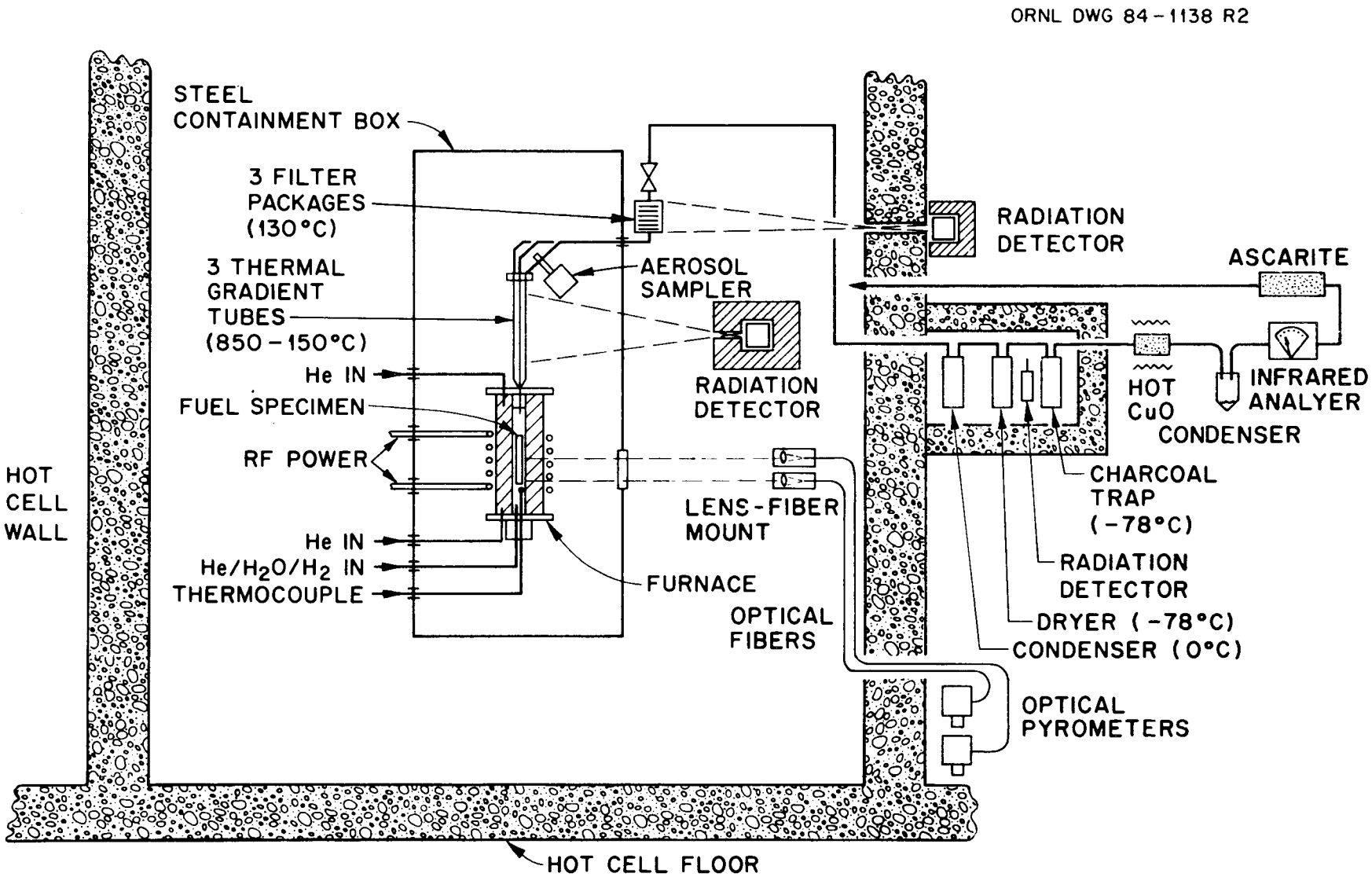

Fig. 1. Vertical fission product release apparatus.

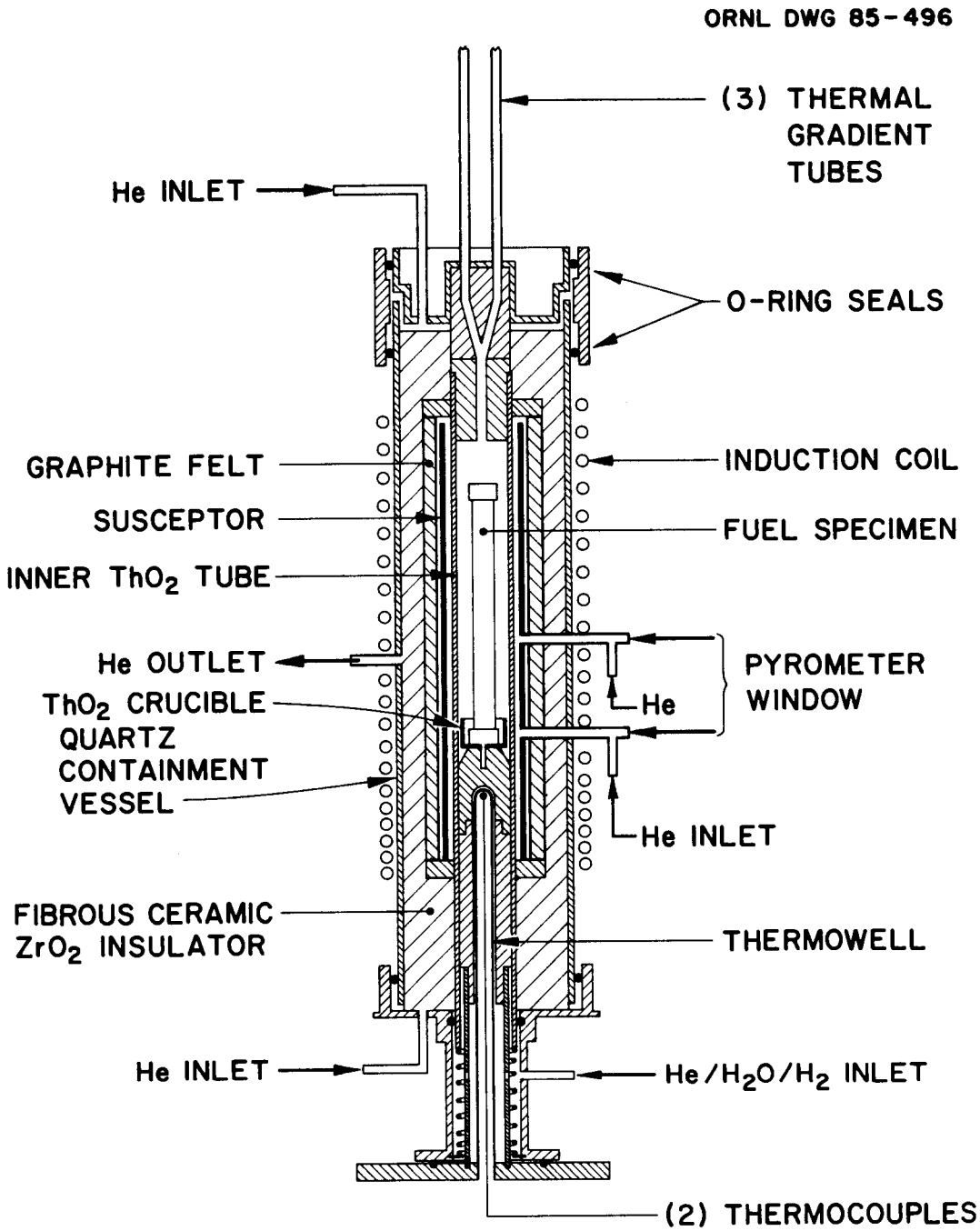

Fig. 2. Vertical fission product release furnace.

the inlet end of the furnace served primarily as a guide during test heatup; the disappearing-filament and two-color pyrometers do not indicate below 800 to 1000°C. The fission product collection apparatus is shown in Fig. 3. Direct weighing of the thermal gradient tube and the filters provides data for the masses of vapor and aerosol released as a function of time. The hot CuO bed converts H_2 to H_2O and CO to CO_2, so that these effluents can be readily measured. (Hydrogen is generated by the reaction of steam with the Zircaloy cladding and with the graphite susceptor, and CO is the other product of the steam–graphite reaction.)

4. DESCRIPTION OF THE FUEL SPECIMEN

The fuel specimen was a 15.2-cm-long section cut from the high-burnup region of rod 08639, assembly No. 1D13, from the Oconee Unit 1 PWR; the irradiation period was Feb. 11, 1975, to Nov. 22, 1979. The burnup was 38.3 MWd/kg, the in-pile release of ^{85}Kr was 4.1% of inventory in the specimen, and the pretest UO_2 grain size was 9.2 μm, as in the specimen used in test HI-5. Like all tests in the HI series, a 1.6-mm-diam hole was drilled through the cladding at midlength to provide a standard leak during heatup.[3]

5. TEST CONDITIONS

This test was intended to investigate fission product release at two temperature levels — 2000 and 2300 K — under strongly oxidizing conditions. The operating conditions are summarized in Table 1, and the temperature history is shown in Fig. 4. The time periods for operation of the three collection trains were Train A: 0 to 72 min, Train B: 72 to 86 min, and Train C: 86 min to end of test, including cooldown. Temperature measurement and control were generally good; the small peak indicating overshoot at the 2000 K level (~55 min) appears to be at least partially the result of oxidation heating, which was near the maximum rate at this time. The 20-min period at 1400 K was included to ensure heatup of ceramics in the outlet end of the furnace before any significant release of fission products had occurred.

6. PRELIMINARY RESULTS

Based on preliminary analyses of the cold charcoal traps (for Kr) and on pretest and posttest gamma analysis of the fuel specimen (for Cs), the total release fractions for Kr and Cs were 52 and 60%, respectively. Compared to measurements of this type in previous tests, this appears to be good agreement. A more precise value for cesium release will be obtained later by summation of the analyses for individual components. The posttest distributions of several fission products in the fuel are shown in Fig. 5; the activity measurements for some isotopes may be low because of uncorrected shielding effects within the fuel. The depression in nuclide concentrations at 12.0 cm is probably the result of fuel shifting during the test. (The top end of the specimen was observed to be off-center when the furnace was opened.)

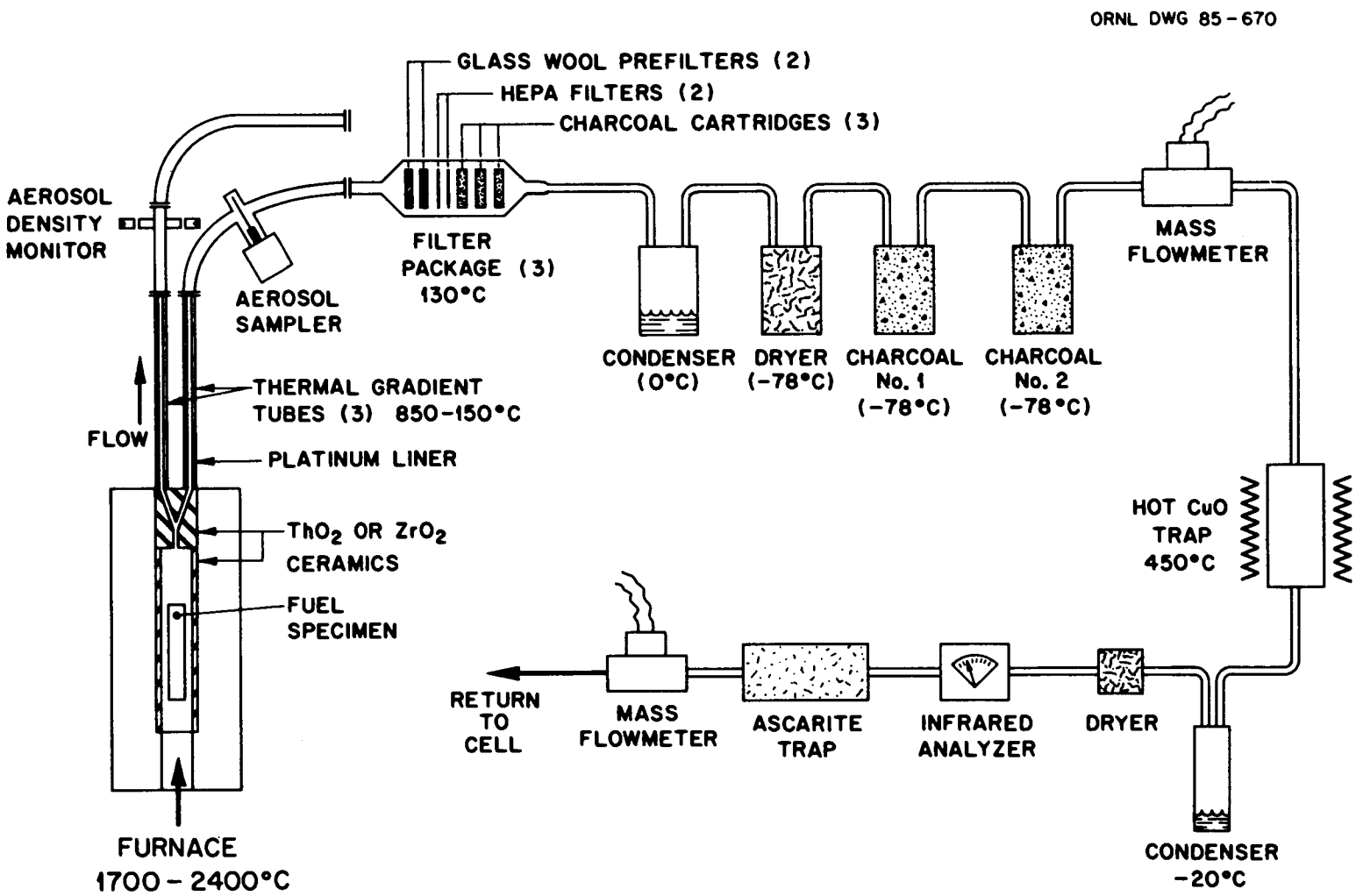

Fig. 3. Fission product and hydrogen collection apparatus.

Table 1. Operating data for test VI-1

Specimen temperature (K):	
At start of heatup ramp	~500
At first 20-min period	1410
At second 20-min period	2020
At third 20-min period	2300
Heatup rate	1/s
Cooldown rate	0.8/s
Time above 2000 K	54 min
Nominal gas flow rate data (L/min at 20°C, 1 bar):[a]	
He purge to thermocouple	0.1
He to recirculation system	0.40
He to steam generator	0.30
Steam into furnace	1.8
Recirculation	1.0
Integral volume flow data (L):	
He purge to thermocouple	15.28
He to recirculation system	52.98
He to steam generator	39.87
Steam into furnace	234
Recirculation	90.8
Total H_2 generated	47.1
H_2 from cladding oxidation[b]	16.2

[a]Measured by mass flowmeters, except steam measured in condenser.

[b]Theoretical value assuming 100% oxidation of Zr.

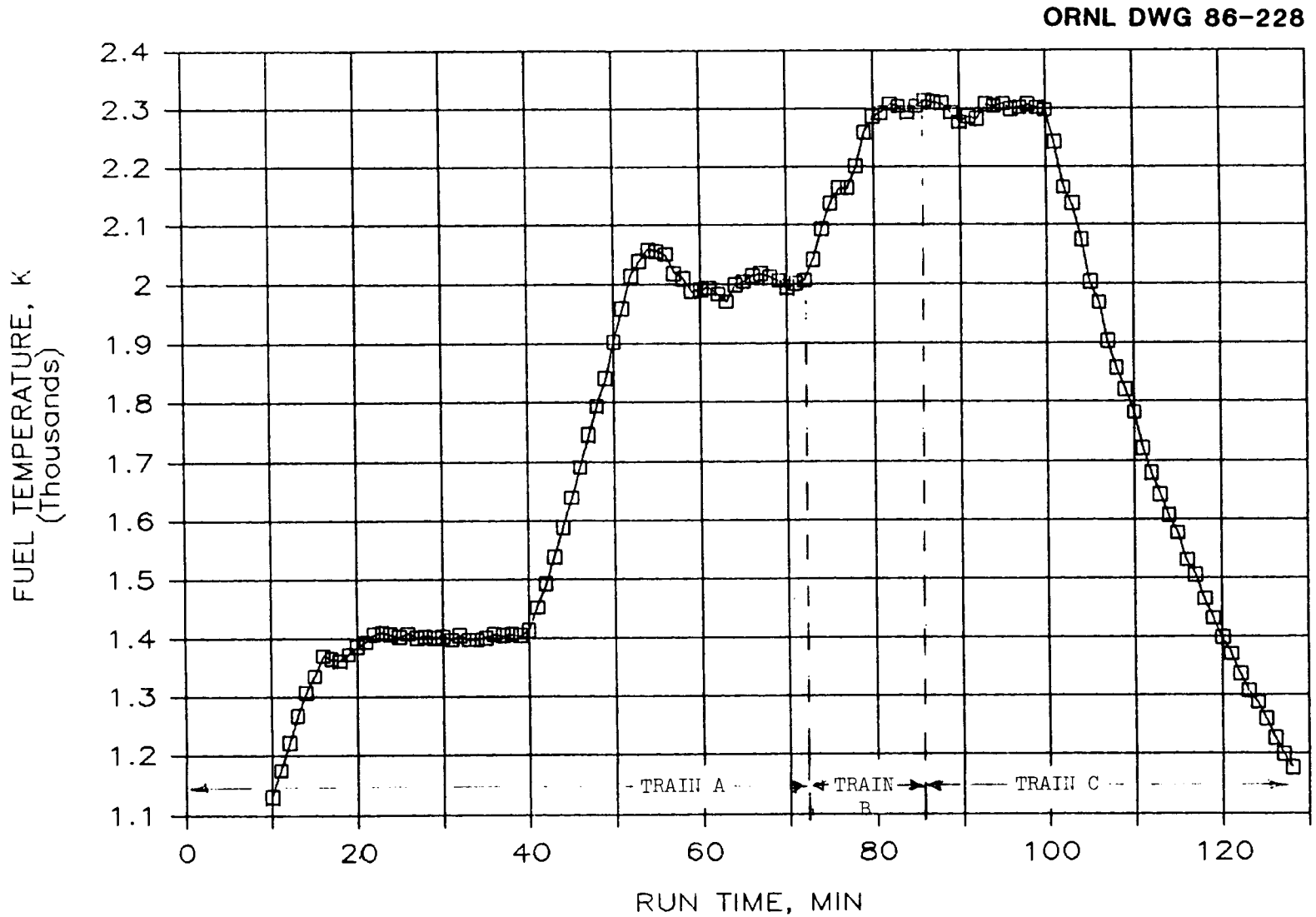

Fig. 4. Temperature history for test VI-1.

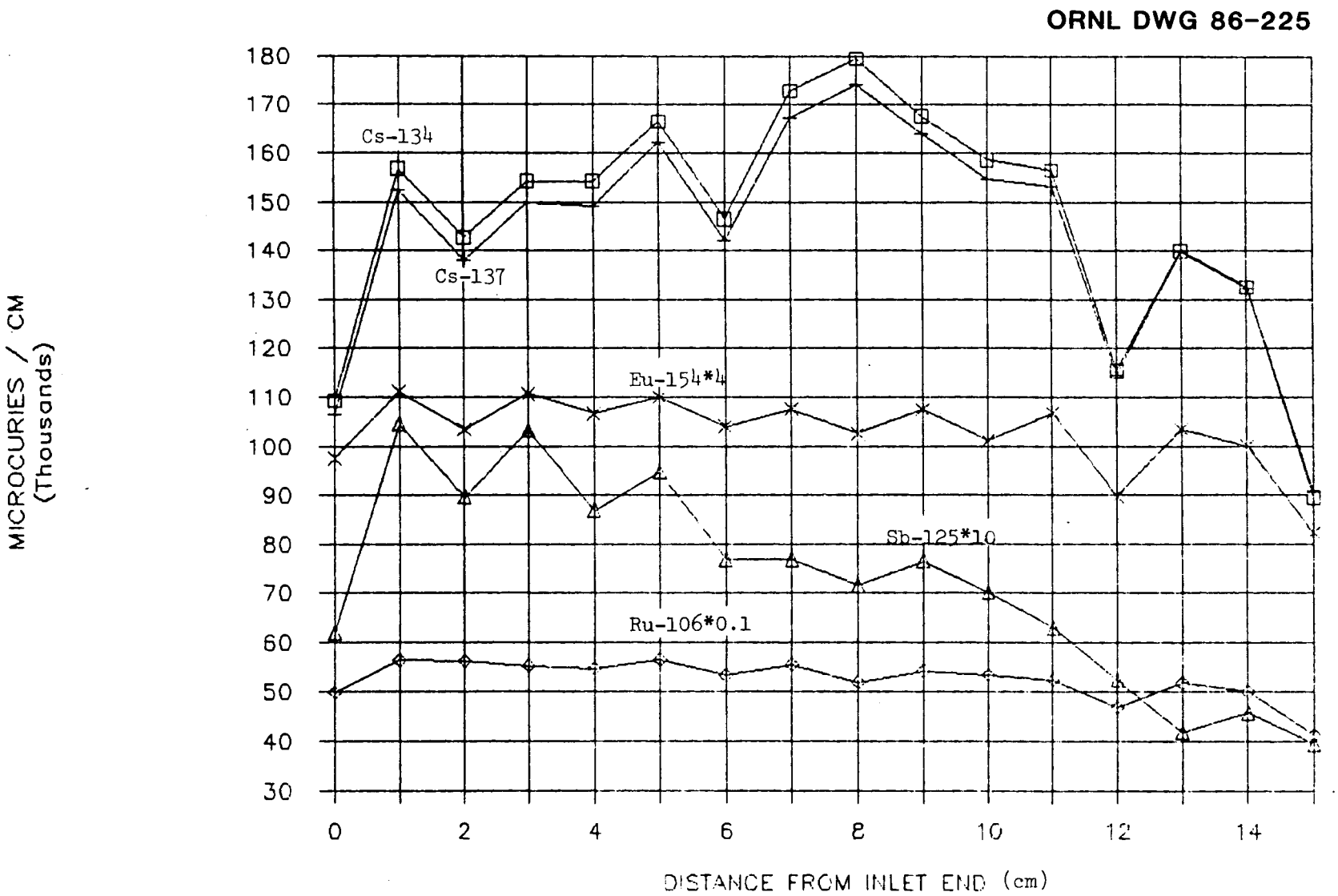

Fig. 5. Distributions of fission products in the fuel after testing.

Direct measurement of the hydrogen generated during the test is shown in Fig. 6; the sharp peak in the rate curve at 54 min is primarily the H_2 resulting from cladding oxidation, while the H_2 evolving after 60 min is primarily the product of graphite susceptor oxidation, with a higher rate indicated at 2300 than at 2000 K. The hydrogen from cladding oxidation in a test of unirradiated fuel, test VT-12, is compared with the amount calculated using generally accepted literature values for the oxidation rates in Fig. 7. As indicated, the experimental values agreed well with the calculated values, falling somewhat below the Baker-Just rate at 1700 K and exceeding the Urbanic-Heidrick rate at 2000 K. Note that the temperature—time history for test VT-12, a rehearsal for test VI-1, was essentially identical to that for test VI-1.

The cesium profiles in the three thermal gradient tubes are illustrated in Figs. 8–10. In addition, small amounts of ^{125}Sb were measured throughout tube C and both ^{125}Sb and ^{110m}Ag were detected at the inlet ends of some tubes. Tubes A and C exhibited the characteristic cesium peaks in the 500–700°C regions, probably indicating the presence of CsI and CsOH deposits. The only unusual features of these curves, compared with those from previous tests, are the small cesium peaks near the outlet ends (at ~250°C) of the tubes. The masses of deposits in the thermal gradient tubes and on the filters are summarized in Table 2. Although visual examination revealed no deposits on Train A filters, visible deposits, including a smooth filter cake on the Train C prefilter, were seen on the other filters. As expected from previous tests, mass release was highest in Train C (2300 K), whereas cesium release reached a maximum in Train A (2000 K) and declined later in the test because of source depletion.

The releases of ^{85}Kr, ^{134}Cs, and ^{137}Cs were monitored continuously as in HI series tests. Higher-than-expected count rates, however, were encountered with the new apparatus geometry, resulting in some saturation of detectors. Consequently, the value of these data from this test appears to be limited; additional shielding will be installed to eliminate this problem in future tests.

7. COMPARISON OF Kr AND Cs RELEASE RATES WITH NUREG-0772

The NUREG-0772 release rate curve for volatile fission products was approximated from 1673 to 2673 K by performing a least-squares analysis for a "1/T correlation." The result was

$$k = 33{,}920 \exp(-28145/T) ,$$

where k is the NUREG-0772 release rate coefficient, min^{-1}, and T is temperature in kelvin. When the above expression was used over the entire time—temperature history of test VI-1, the predicted release was 99.3%, obviously much greater than the observed release for Kr and Cs. When the above rate expression was divided by six, the predicted release was 56.6%, very close to the observed amounts. This average release rate is shown in Fig. 11 as a dotted line from 2000 to 2300 K (1725 to 2025°C). This preliminary average release rate, about a factor of six below the NUREG-0772 curve, is in line with most of the results from the HI test series: HI-1, -3, -4, and -5.

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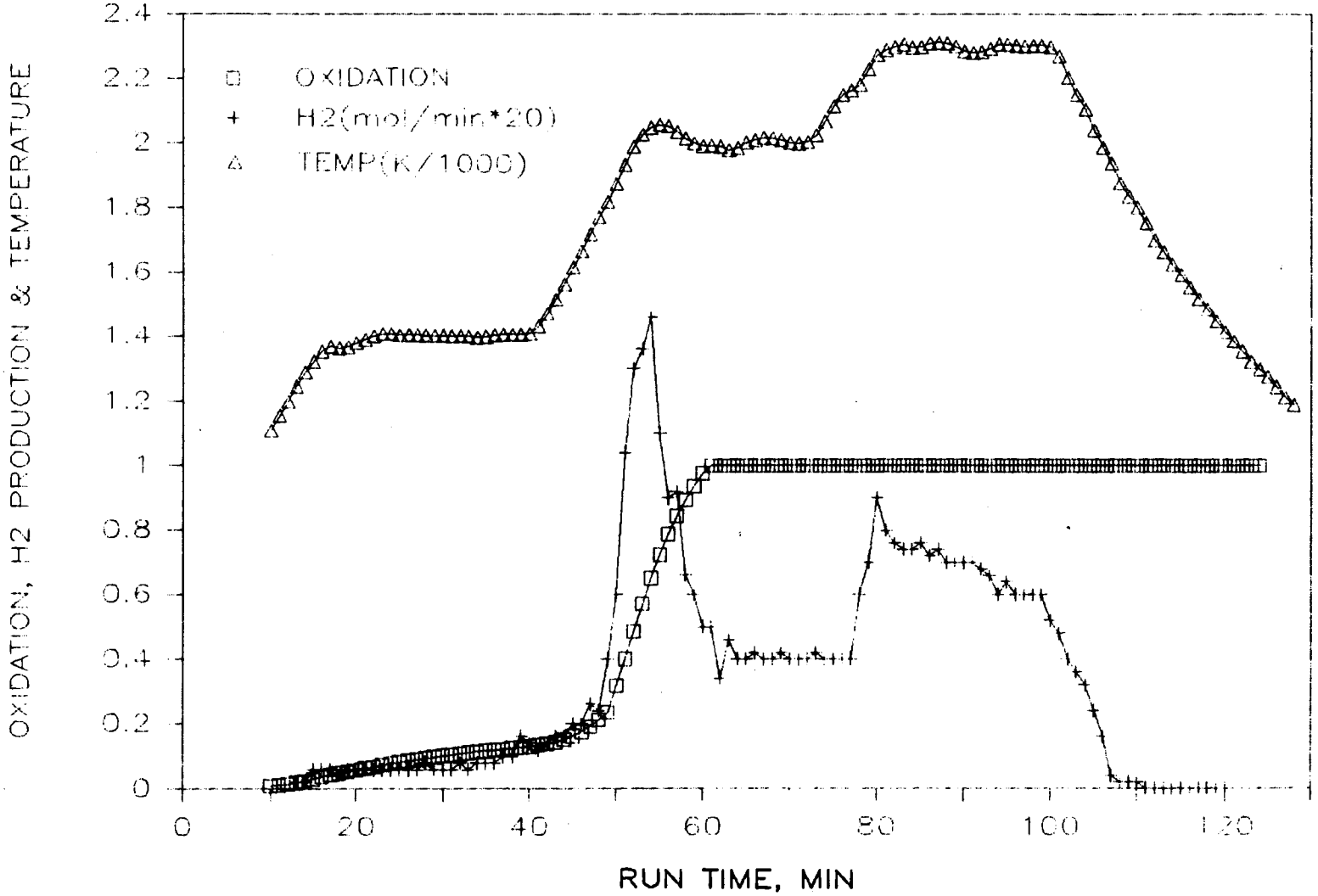

Fig. 6. Cladding oxidation and hydrogen production in test VI-1 as a function of time (temperature).

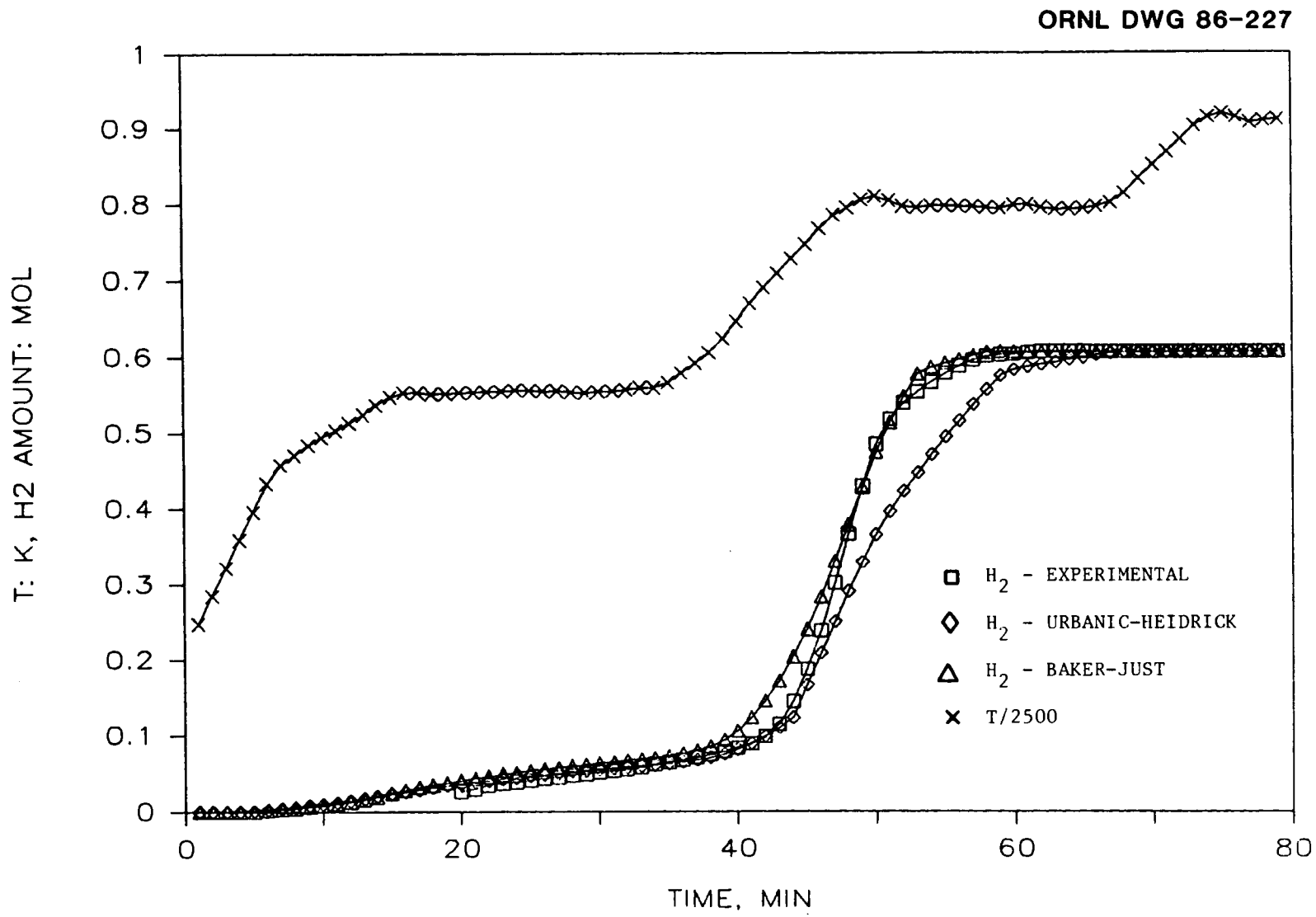

Fig. 7. Comparison of measured and calculated hydrogen production in a test simulating VI-1.

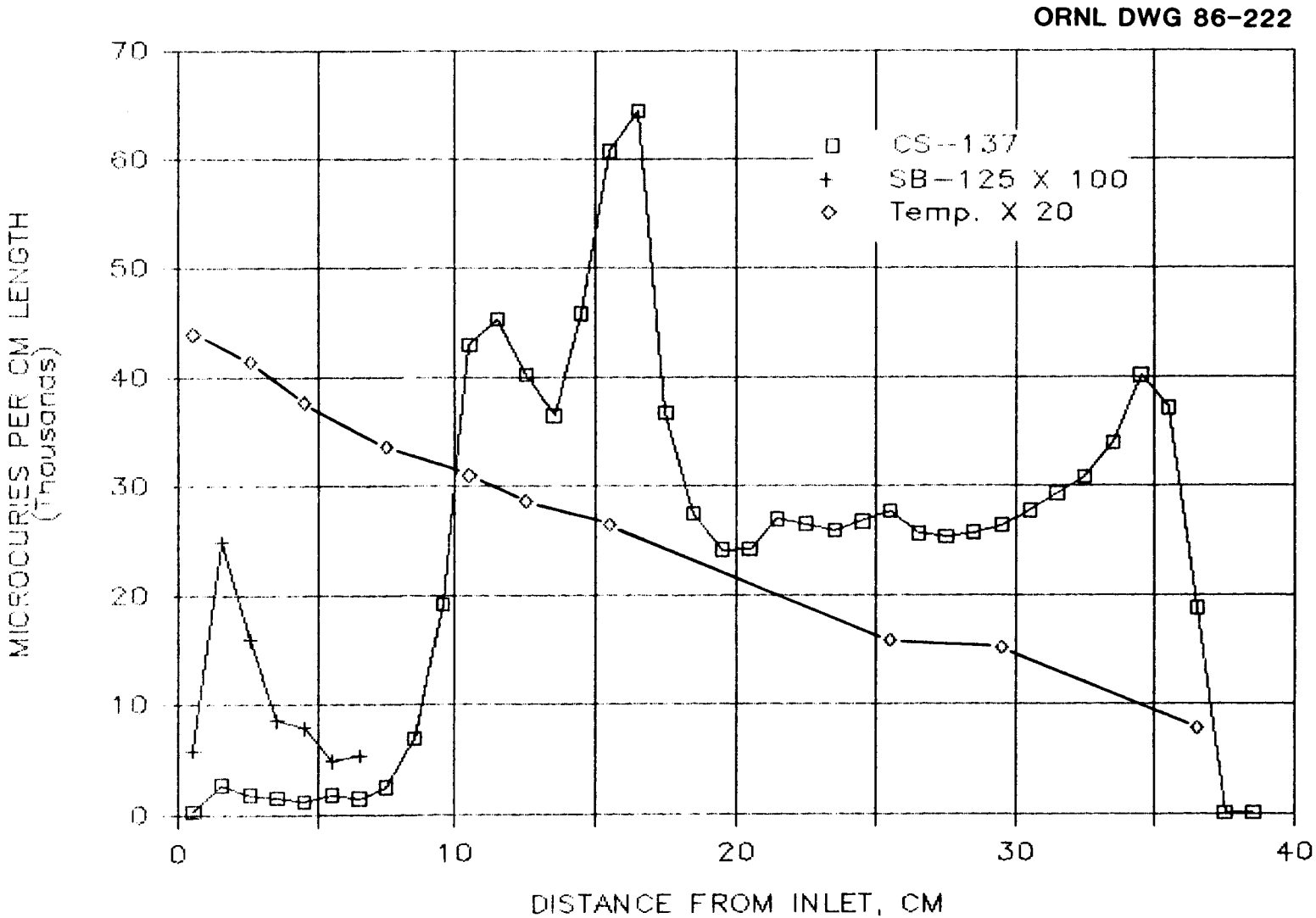

Fig. 8. Distribution of cesium and antimony in thermal gradient tube A.

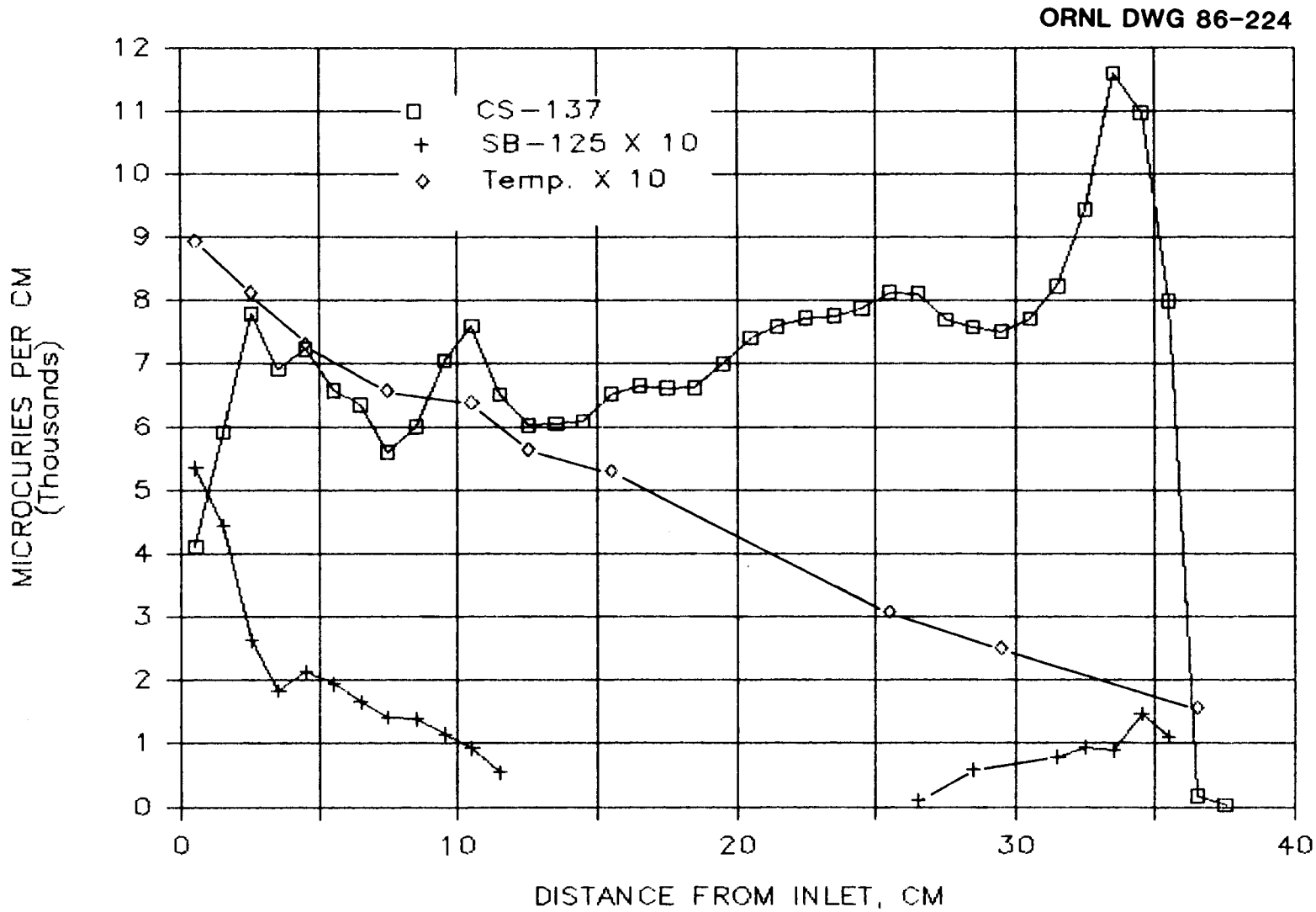

Fig. 9. Distribution of cesium and antimony in thermal gradient tube B.

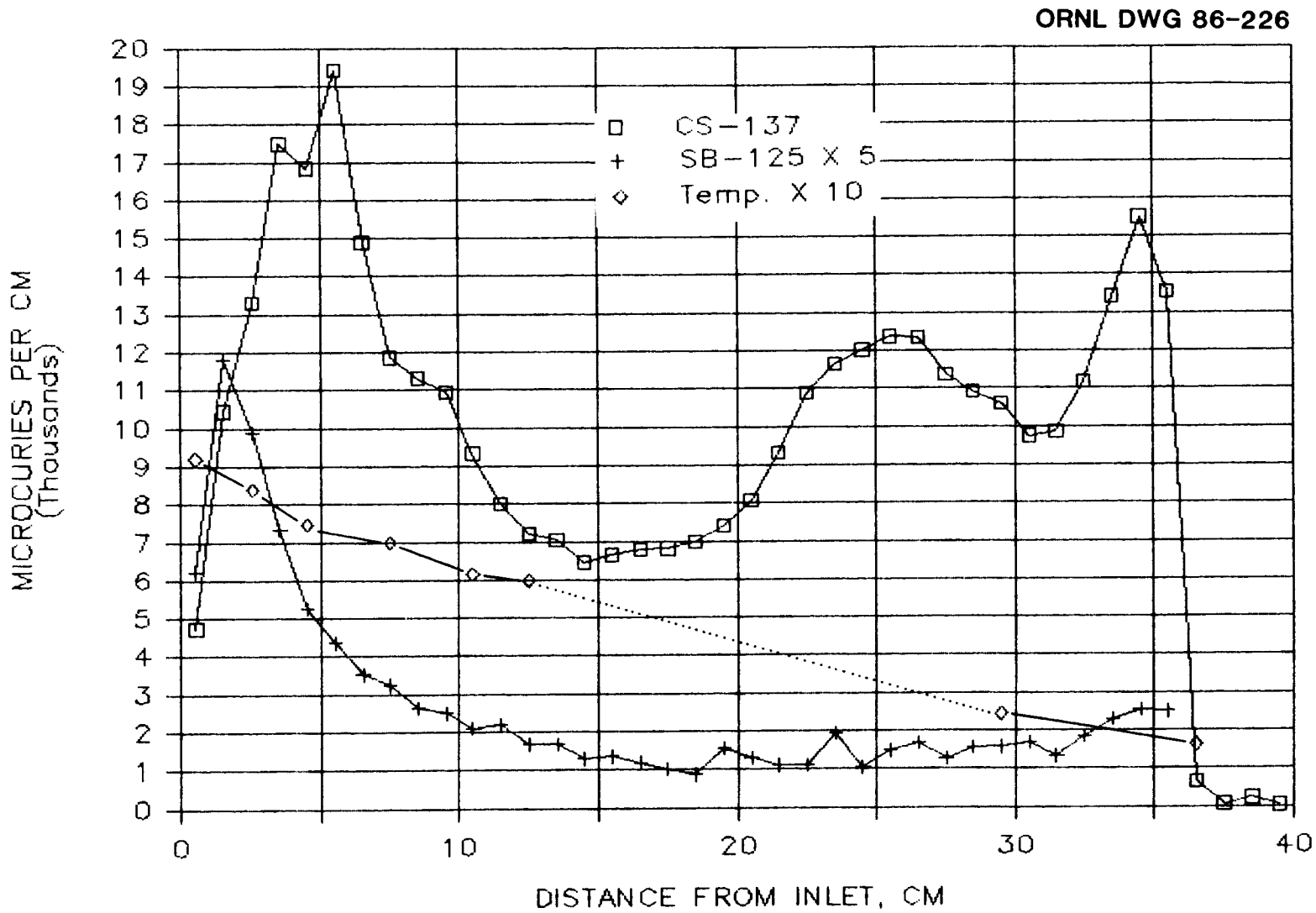

Fig. 10. Distribution of cesium and antimony in thermal gradient tube C.

Table 2. Vapor and aaerosol deposits in test VI-1

	Weight of deposits (g)[a]		
	Train A	Train B	Train C
Thermal gradient tube (TGT)	0.078	0.101	0.225
Filters:			
Prefilter 1	0.134	0.157	0.213
Prefilter 2	0.050	0.030	0.029
HEPA 1	0.018	0.007	0.008
HEPA 2	0.018	0.004	0.008
Total filters	0.220	0.198	0.258
TGT and filters	0.298	0.299	0.483
Total experiment			1.080

[a]Precision ±0.003 g.

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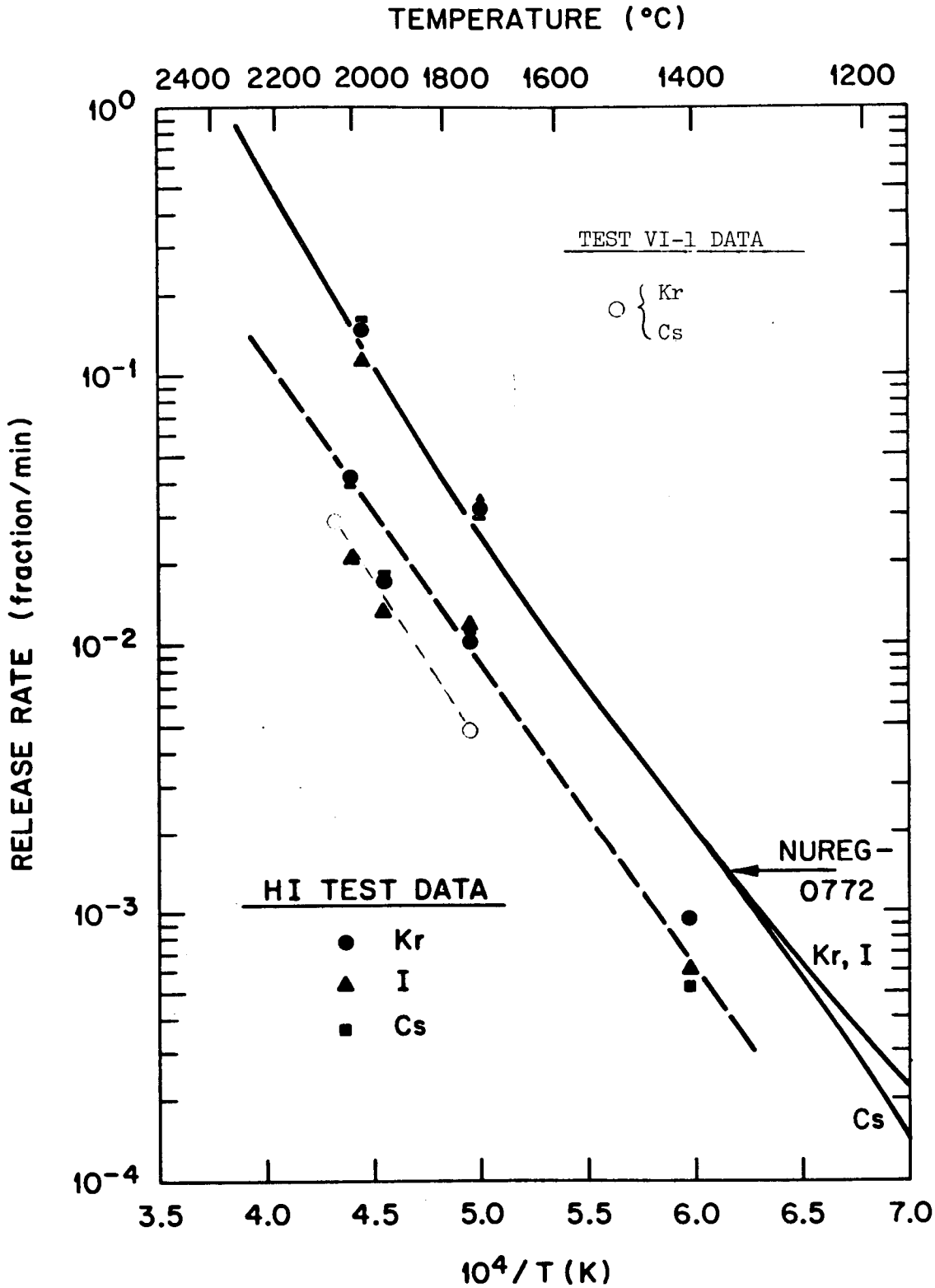

Fig. 11. Comparison of release rate coefficients for test VI-1 with data from HI tests.

A valid criticism of the NUREG-0772 release model is that it is too simple to cover the wide variety of fuel types, burnups, heatup and cool-down rates, time at temperature, chemical environment, etc., that can actually exist in a reactor accident scenario. Very preliminary measurements of cesium collected during test VI-1 indicate that more was collected in Train A than in Trains B or C. Conversely, the NUREG-0772 release model with rates reduced by a factor of six predicts 10% release to Train A, 21% to Train B, and 25% to Train C. This disagreement with NUREG-0772 kinetics has prompted us to look again at the change in release with time (or more correctly, the change with amount released). A first attempt at a new, simple empirical model gave 25% release to Train A, 16% to Train B, and 16% to Train C. This is the same distribution that would be obtained using the Booth model (diffusion from an equivalent sphere) and an activation energy of 55,920 cal/mol (derived from the NUREG-0772 curve). Further analysis of these results is in progress.

A preliminary calculation was performed with the VI-1 time—temperature history using the EPRI/IDCOR "Steam Oxidation" fission product release model. The calculation gave 100% release with the distribution as follows: 99% to Train A, 1% to Train B, and nothing remaining for Train C. The steam oxidation model release rates decrease with time, but the initial release rate appears to be much too high.

8. CONCLUSIONS

By any standard, this was a very successful initial test in a new facility. Most of the test equipment, especially that used for temperature measurement and control, operated quite well. Observed and calculated values for hydrogen production during the test were in good agreement. The preliminary data for krypton and cesium release were consistent with data from previous tests in the HI series. Following completion of data collection and analysis from this test, a comprehensive data summary report will be issued. The available data indicate that this test project should provide information valuable in developing an improved fission product release and behavior model for LWR fuel under accident conditions.

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